

APPENDIX B, Part 2

DECLARATION OF TRANSLATOR

I, Maho KASEKI, c/o the Inoue & Associates of 3rd Floor, Akasaka Habitation Building, 3-5, Akasaka 1-chome, Minato-ku, Tokyo, Japan do solemnly and sincerely declare that I am well acquainted with the Japanese and English languages and that I have executed with the best of my ability this translation into English of the Certified Copy of Japanese Patent Application No. 2003-153159 and believe that the translation is true and correct.

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

April 19, 2005  
(Date)

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[Title of the Invention] Conductive Masterbatch and Conductive Resin Composition

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[List of Materials Being Submitted]

[Name of Material]	Specification	1
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[Name of Material]	Drawing	1
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[Name of Material]	Abstract	1
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[Necessity or Non-Necessity of Proof] Necessary

[Name of Document] Specification

[Title of the Invention] Conductive masterbatch and conductive resin composition

[Scope of Claims for Patent]

[Claim 1] A conductive masterbatch comprising a polyamide and a conductive carbon black, wherein a part of said conductive carbon black is present in the form of 1 to 100 agglomerated particles each independently having a major axis of 20 to 100  $\mu\text{m}$ , as observed under an optical microscope with respect to a contiguous area of 3  $\text{mm}^2$ .

[Claim 2] The conductive masterbatch according to claim 1, wherein 1 to 50 agglomerated particles of said carbon black, each independently having a major axis of 20 to 100  $\mu\text{m}$ , are present, as observed under an optical microscope with respect to a contiguous area of 3  $\text{mm}^2$ .

[Claim 3] The conductive masterbatch according to claim 1, wherein 2 to 40 agglomerated particles of said carbon black, each independently having a major axis of 20 to 100  $\mu\text{m}$ , are present, as observed under an optical microscope with respect to a contiguous area of 3  $\text{mm}^2$ .

[Claim 4] The conductive masterbatch according to claim 1, wherein 2 to 30 agglomerated particles of said carbon black, each independently having a major axis of 20 to 100  $\mu\text{m}$ , are present, as observed under an optical microscope with respect to a contiguous area of 3  $\text{mm}^2$ .

[Claim 5] The conductive masterbatch according to any one of claims 1 to 5, wherein the DBP oil absorption of said conductive carbon black is 250 ml or more of 100 g of the carbon black.

[Claim 6] The conductive masterbatch according to any one of claims 1 to 5, which is in the form of pellets.

[Claim 7] The conductive masterbatch according to claim 6, wherein the surface roughness of each of the pellets is in the range of from 0.3 to 2.0  $\mu\text{m}$  in terms of the central line average roughness (Ra) value as measured by a surface roughness gauge.

[Claim 8] The conductive masterbatch according to claim 6, wherein the surface roughness of each of the pellets is in the range of from 0.4 to 1.5  $\mu\text{m}$  in terms of the central line average roughness (Ra) value as measured by a surface roughness gauge.

[Claim 9] The conductive masterbatch according to claim 7 or 8, wherein each of said pellets has a diameter of from 1.5 to 3.5 mm and a length of from 2.0 to 3.5 mm.

[Claim 10] The conductive masterbatch according to any one of claims 1 to 9, wherein the amount of said conductive carbon black in the conductive masterbatch is in the range of from 5 to 40 % by weight, based on the weight of the masterbatch.

[Claim 11] The conductive masterbatch according to claim 10, wherein the amount of said conductive carbon black is in

the range of from 6 to 10 % by weight, based on the weight of the masterbatch.

[Claim 12] A conductive resin composition comprising a polyamide, a polyphenylene ether and a conductive carbon black, which is produced by using the conductive masterbatch of any one of claims 1 to 11.

[Claim 13] The conductive resin composition according to claim 12, wherein the amount of said conductive carbon black is in the range of from 0.2 to 5 parts by weight, relative to 100 parts by weight of the total of the components of the conductive resin composition exclusive of the conductive carbon black.

[Claim 14] The conductive resin composition according to claim 12 or 13, which is for use in the production of an automobile outer panel.

[Claim 15] An injection molded article comprising the conductive resin composition of any one of claims 12 to 14.

[Claim 16] A molded article for use in the production of an automobile outer panel, which comprises the conductive resin composition of any one of claims 12 to 14.

[Claim 17] An automobile fender, which comprises the conductive resin composition of any one of claims 12 to 14.

[Claim 18] An automobile fender, which is an injection molded article comprising the conductive resin composition of any one of claims 12 to 14 and having a surface roughness of from 0.05 to 1  $\mu\text{m}$ .

[Claim 19] A method for producing a conductive resin composition comprising a polyamide, a polyphenylene ether and a conductive carbon black, which comprises the following steps:

(1) producing a conductive masterbatch comprising a polyamide and a conductive carbon black, wherein a part of said conductive carbon black is present in the form of an agglomerated particle having a major axis of 20 to 100  $\mu\text{m}$ , and

(2) adding the produced conductive masterbatch to a molten polyphenylene ether.

[Claim 20] The method according to claim 19, wherein a polyamide is added to the molten polyphenylene ether, simultaneously with the addition of the conductive masterbatch.

[Claim 21] The method according to claim 19 or 20, wherein, as said conductive masterbatch, the conductive masterbatch of any one of claims 1 to 11 is used.

[Detailed Description of the Invention]

[0001]

[Field of the Invention]

The present invention relates to a masterbatch used for producing a resin composition having excellent impact resistance and excellent conductivity, which is advantageous in that, when the resin composition is molded into a large article (such as an automobile outer panel) and the resultant molded article is then subjected to an electrostatic coating, the resin composition is unlikely to suffer heat distortion and a lowering of

the impact resistance, wherein excellent coating can be formed on such a molded article due to the excellent conductivity of the resin composition. The present invention is also concerned with a resin composition and molded article which are produced using the above-mentioned masterbatch; and a method for producing the above-mentioned resin composition or molded article.

The resin composition of the present invention can be used in a wide variety of fields, such as electric and electronic parts, parts of office automation machines, automobile parts and other mechanical parts. Especially, the resin composition can be advantageously used for producing an automobile outer panel part which can be electrostatically coated.

[0002]

[Prior Art]

Polyphenylene ethers not only have excellent mechanical and electrical properties and excellent heat resistance, but also have excellent dimensional stability. Therefore, polyphenylene ethers have been used in a wide variety of fields. However, the moldability of a polyphenylene ether is poor. For improving the moldability of a polyphenylene ether, patent document 1 proposes a technique in which a polyamide is added to a polyphenylene ether to thereby obtain a polyamide-polyphenylene ether alloy. Nowadays, polyamide-polyphenylene ether alloys are used in a very wide variety of fields.

[0003]

Recently, use of a conductivity-imparted polyamide-polyphenylene ether alloy is rapidly expanding in the fields of electrostatically coatable automobile outer panels (such as a fender and a door panel). The materials used for an automobile outer panel are required to have various good properties, such as a conductivity sufficient for electrostatic coating, impact resistance, heat resistance and melt-fluidity.

[0004]

With respect to the method for imparting a conductivity to a polyamide-polyphenylene ether alloy, for example, patent document 2 discloses a technique to lower the surface resistance of a polyamide-polyphenylene ether alloy by a method in which carbon black is caused to be contained mainly in the polyamide phase of the polyamide-polyphenylene ether alloy, or by a method in which carbon black is uniformly dispersed in a polyamide, followed by mixing thereof with a polyphenylene ether. There have generally been known a method for uniformly dispersing fine particles (such as particles of carbon black) in a high viscosity substance, and products obtained by such a method (see, for example, non-patent document 1).

[0005]

Patent document 3 discloses a technique in which a polyamide and a polyphenylene ether are compatibilized with each other in advance, followed by addition of carbon black. Patent document 4 describes that, in the production of a composition

containing a polyamide, a polyphenylene ether, carbon black and a compatibility agent, by adjusting the amounts of these components and using a polyphenylene ether and a polyamide which have specific relative viscosities, it becomes possible to improve the conductivity and processability of the composition.

[0006]

Patent document 5 discloses a technique in which a conductive carbonaceous filler (KB) is caused to be present in a particulate form in the polyphenylene ether phase of a polyamide/polyphenylene ether resin composition. In this patent document, the structure of the resin composition is observed under a transmission electron microscope with a magnification as high as 20,000, but the observation is made only to confirm the presence of KB particles (size: around 50 nm) which are much smaller than 20  $\mu\text{m}$ . Further, the impact resistance of the resin composition is unsatisfactory. In this patent document, the resin composition is produced using a masterbatch; however, there is no description about the specific features of the masterbatch.

[0007]

The resin compositions and molded articles obtained by the above-mentioned conventional techniques are likely to suffer heat distortion. Further, conventionally, it has been impossible to produce a resin composition which is improved with respect to all of the conductivity, impact resistance and heat

distortion resistance. Especially, the conventional resin compositions are not suitable for use in applications, such as the production of an article (such as an automobile outer panel) which is large in size and which needs to be used in a processing involving an electrostatic coating and needs to have high heat distortion resistance, high impact resistance and conductivity sufficient for electrostatic coating. Therefore, the development of a new technique has been desired in the art.

[0008]

[Patent document 1]

Examined Japanese Patent Application Publication No.

Sho 45-997 (page 1, claims)

[Patent document 2]

Unexamined Japanese Patent Application Laid-Open

Specification No. Hei 2-201811 (page 1, claims 1 and 2)

[Patent document 3]

Unexamined Japanese Patent Application Laid-Open

Specification No. Hei 8-48869 (page 2, claims 7 to 10)

[Patent document 4]

Unexamined Japanese Patent Application Laid-Open

Specification No. Hei 4-300956 (page 3, paragraph

[0005])

[Patent document 5]

International Patent Application Publication WO

01/81473

[Non-patent document 1]

"Kagakugijutsushi (Journal of Chemical Technology) MOL",

pages 41-46, November 1987

[0009]

[Problems to Be Solved by the Invention]

It is an object of the present invention to provide a resin composition having excellent impact resistance and excellent conductivity, which is advantageous in that, when the resin composition is molded into a large article (such as an automobile outer panel) and the resultant molded article is then subjected to an electrostatic coating, the resin composition is unlikely to suffer heat distortion and a lowering of the impact resistance, wherein excellent coating can be formed on such a molded article due to the excellent conductivity of the resin composition. It is another object of the present invention to provide a conductive masterbatch used for producing the above-mentioned resin composition. It is still another object of the present invention to provide a method for producing the above-mentioned resin composition. It is still another ob-

ject of the present invention to provide a molded article produced using the above-mentioned resin composition.

[0010]

[Means to Solve the Problem]

The present inventors have made extensive and intensive studies with a view toward solving the above-mentioned problems. As a result, it has surprisingly been found that a resin composition produced using a specific masterbatch not only is unlikely to suffer heat distortion or a lowering of the impact resistance, but also has a conductivity sufficient for electrostatic coating. The above-mentioned specific masterbatch comprises a conductive carbon black, wherein the conductive carbon black is present in the form of 1 to 100 agglomerated particles each independently having a major axis of 20 to 100  $\mu\text{m}$ , as observed under an optical microscope with respect to a contiguous area of  $3 \text{ mm}^2$ . The present invention has been completed, based on this finding.

[0011]

The present invention relates to a masterbatch comprising a polyamide and a conductive carbon black, wherein a part of the conductive carbon black is present in the form of agglomerated particles. The present invention is also concerned with a conductive resin composition produced using the above-mentioned masterbatch, which not only is unlikely to suffer heat distortion, but also has both impact resistance and a conductivity

which is comparable or superior to that of a conventional material (i.e., conductivity sufficient for the resin composition to be electrostatically coated). Further, the present invention also relates to an injection molded article (especially a molded article for an automobile outer panel) comprising the above-mentioned resin composition. Further, the present invention also relates to a method for producing the resin composition.

According to the present invention, the following are provided.

[0012]

(1) A conductive masterbatch comprising a polyamide and a conductive carbon black, wherein a part of the conductive carbon black is present in the form of 1 to 100 agglomerated particles each independently having a major axis of 20 to 100  $\mu\text{m}$ , as observed under an optical microscope with respect to a contiguous area of 3  $\text{mm}^2$ .

(2) The conductive masterbatch according to item (1) above, wherein 1 to 50 agglomerated particles of the carbon black, each independently having a major axis of 20 to 100  $\mu\text{m}$ , are present, as observed under an optical microscope with respect to a contiguous area of 3  $\text{mm}^2$ .

(3) The conductive masterbatch according to item (1) above, wherein 2 to 40 agglomerated particles of the carbon black, each independently having a major axis of 20 to 100  $\mu\text{m}$ , are

present, as observed under an optical microscope with respect to a contiguous area of 3 mm<sup>2</sup>.

(4) The conductive masterbatch according to item (1) above, wherein 2 to 30 agglomerated particles of the carbon black, each independently having a major axis of 20 to 100 µm, are present, as observed under an optical microscope with respect to a contiguous area of 3 mm<sup>2</sup>.

[0013]

(5) The conductive masterbatch according to any one of items (1) to (5) above, wherein the DBP oil absorption of the conductive carbon black is 250 ml or more of 100 g of the carbon black.

(6) The conductive masterbatch according to any one of items (1) to (5) above, which is in the form of pellets.

(7) The conductive masterbatch according to item (6) above, wherein the surface roughness of each of the pellets is in the range of from 0.3 to 2.0 µm in terms of the central line average roughness (Ra) value as measured by a surface roughness gauge.

(8) The conductive masterbatch according to item (6) above, wherein the surface roughness of each of the pellets is in the range of from 0.4 to 1.5 µm in terms of the central line average roughness (Ra) value as measured by a surface roughness gauge.

[0014]

(9) The conductive masterbatch according to item (7) or (8) above, wherein each of the pellets has a diameter of from 1.5 to 3.5 mm and a length of from 2.0 to 3.5 mm.

(10) The conductive masterbatch according to any one of items (1) to (9) above, wherein the amount of the conductive carbon black in the conductive masterbatch is in the range of from 5 to 40 % by weight, based on the weight of the masterbatch.

(11) The conductive masterbatch according to item (10) above, wherein the amount of the conductive carbon black in the conductive masterbatch is in the range of from 6 to 10 % by weight, based on the weight of the masterbatch.

[0015]

(12) A conductive resin composition comprising a polyamide, a polyphenylene ether and a conductive carbon black, which is produced by using the conductive masterbatch of any one of items (1) to (11) above.

(13) The conductive resin composition according to item (12) above, wherein the amount of the conductive carbon black is in the range of from 0.2 to 5 parts by weight, relative to 100 parts by weight of the total of the components of the conductive resin composition exclusive of the conductive carbon black.

(14) The conductive resin composition according to item (12) or (13) above, which is for use in the production of an

automobile outer panel.

[0016]

(15) An injection molded article comprising the conductive resin composition of any one of items (12) to (14) above.

(16) A molded article for use in the production of an automobile outer panel, which comprises the conductive resin composition of any one of items (12) to (14) above.

(17) An automobile fender, which comprises the conductive resin composition of any one of items (12) to (14) above.

(18) An automobile fender, which is an injection molded article comprising the conductive resin composition of any one of items (12) to (14) above and having a surface roughness of from 0.05 to 1  $\mu\text{m}$ .

[0017]

(19) A method for producing a conductive resin composition comprising a polyamide, a polyphenylene ether and a conductive carbon black, which comprises the following steps:

(1) producing a conductive masterbatch comprising a polyamide and a conductive carbon black, wherein a part of the conductive carbon black is present in the form of an agglomerated particle having a major axis of 20 to 100  $\mu\text{m}$ , and

(2) adding the produced conductive masterbatch to a molten polyphenylene ether.

(20) The method for producing a conductive resin composition according to item (19) above, wherein, a polyamide is

added to the molten polyphenylene ether, simultaneously with the addition of the conductive masterbatch.

(21) The method according to item (19) or (20) above, wherein, as the masterbatch, the masterbatch of any one of items (1) to (11) above is used.

[0018]

[Mode for Carrying Out the Invention]

Hereinbelow, the components of the conductive resin composition of the present invention will be described in detail.

<Polyamide>

With respect to the type of the polyamide which can be used in the present invention, there is no particular limitation so long as it is a polymer having amide  $\{-\text{NH}-\text{C}(=\text{O})-\}$  linkages in a main chain thereof.

In general, a polyamide is obtained by a ring opening polymerization of a lactam, a condensation polymerization of a diamine and a dicarboxylic acid, a condensation polymerization of an aminocarboxylic acid, or the like. However, in the present invention, the method for obtaining a polyamide is not limited to these examples.

[0019]

Examples of diamines mentioned above include aliphatic diamines, alicyclic diamines and aromatic diamines. Specifically, there can be mentioned tetramethylenediamine, hexamethylenediamine, undecamethylenediamine, dodecamethylenediamine,

tridecamethylenediamine, 2,2,4-trimethylhexamethylenediamine, 2,4,4-trimethylhexamethylenediamine, 5-methylnonamethylenediamine, 1,3-bisaminomethylcyclohexane, 1,4-bisaminomethylcyclohexane, m-phenylenediamine, p-phenylenediamine, m-xylylenediamine and p-xylylenediamine.

[0020]

Examples of dicarboxylic acids include aliphatic dicarboxylic acids, alicyclic dicarboxylic acids and aromatic dicarboxylic acids. Specifically, there can be mentioned adipic acid, suberic acid, azelaic acid, sebacic acid, dodecanoic diacid, 1,1,3-tridecanoic diacid, 1,3-cyclohexane dicarboxylic acid, terephthalic acid, isophthalic acid, naphthalene dicarboxylic acid and a dimer acid.

[0021]

Specific examples of lactams include  $\epsilon$ -caprolactam, enanthlactam and  $\omega$ -laurocaprolactam.

Further, specific examples of aminocarboxylic acids include  $\epsilon$ -aminocaproic acid, 7-aminoheptanoic acid, 8-aminoctanoic acid, 9-aminononanoic acid, 11-aminoundecanoic acid, 12-aminododecanoic acid and 13-aminotridecanoic acid.

[0022]

In the present invention, the polyamide may either be a homopolymer obtained by homopolymerizing any one of the above-mentioned compounds (i.e., lactams, diamines, dicarboxylic acids and  $\omega$ -aminocarboxylic acids), or a copolymer ob-

tained by subjecting a mixture of at least two types of the above-mentioned compounds to a condensation polymerization.

In the present invention, it is also preferred to use a polyamide obtained by a method in which at least one of the above-mentioned compounds (i.e., lactams, diamines, dicarboxylic acids and  $\omega$ -aminocarboxylic acids) is polymerized in a polymerization reactor to thereby obtain a low molecular weight oligomer, and the obtained oligomer is subjected to further polymerization in an extruder or the like, to thereby obtain a high molecular weight polymer.

[0023]

Examples of polyamides which can be advantageously used in the present invention include polyamide 6, polyamide 6,6, polyamide 4,6, polyamide 11, polyamide 12, polyamide 6,10, polyamide 6,12, polyamide 6/6,6, polyamide 6/6,12, polyamide 6/MXD (m-xylylenediamine), polyamide 6,T, polyamide 6,I, polyamide 6/6,T, polyamide 6/6,I, polyamide 6,6/6,T, polyamide 6,6/6,I, polyamide 6/6,T/6,I, polyamide 6,6/6,T/6,I, polyamide 6/12/6,T, polyamide 6,6/12/6,T, polyamide 6/12/6,I and polyamide 6,6/12/6,I. Further, it is also possible to use a polyamide which is obtained by copolymerizing a plurality of different polyamides using an extruder or the like.

Preferred examples of polyamides include polyamide 6, polyamide 6,6, a polyamide 6/6,6, and a mixture thereof.

[0024]

The number average molecular weight of the polyamide used in the present invention is preferably 5,000 to 100,000, more preferably 10,000 to 30,000.

The polyamide used in the present invention is not limited to those which are exemplified above, and may be a mixture of a plurality of polyamides having different molecular weights. For example, the polyamide may be a mixture of a low molecular weight polyamide having a number average molecular weight of less than 15,000 and a high molecular weight polyamide having a number average molecular weight of 150,000 or more.

[0025]

The terminal groups of the polyamide participate in a reaction with a polyphenylene ether. A polyamide generally has an amino group and a carboxyl group as terminal groups thereof. In general, when the carboxyl group concentration of a polyamide resin is increased, the impact resistance of such a polyamide becomes lowered whereas the melt-fluidity of the polyamide becomes improved. On the other hand, when the amino group concentration of a polyamide resin is increased, the impact resistance of such a polyamide becomes improved whereas the melt-fluidity of the polyamide becomes lowered.

In the present invention, the concentration ratio of the amino group to the carboxyl group (amino group/carboxyl group ratio) is preferably 9/1 to 1/9, more preferably 8/2 to 1/9, still more preferably 6/4 to 1/9.

[0026]

The concentration of the terminal amino group of the polyamide is preferably at least 10 milliequivalents, more preferably at least 30 milliequivalents, per kg of the polyamide.

Any conventional methods can be used to control the amounts of the terminal groups of the polyamides. For example, there can be mentioned a method in which a diamine, dicarboxylic acid or monocarboxylic acid is added to the reaction system of a polymerization for producing a polyamide so as to obtain a polyamide having a desired terminal amino group concentration, and a method in which two or more different polyamides having different terminal group ratios are mixed together.

[0027]

Further, for improving the heat stability of a polyamide, a metal compound stabilizer as described in Unexamined Japanese Patent Application Laid-Open Specification No. Hei 1-163262 may be used.

Among the conventional metal compound stabilizers, especially preferred are CuI, CuCl<sub>2</sub>, copper acetate and cerium stearate. It is also preferred to use halogen salts of alkali metals, such as potassium iodide and potassium bromide. These metal compounds stabilizers can be used individually or in combination.

It is preferred that the metal compound stabilizer and/or the halogen salt of alkali metal is added to the polyamide in

an amount of 0.001 to 1 part by weight, relative to 100 parts by weight of the polyamide.

Further, any of other conventional additives for a polyamide can be also added to the polyamide. Such additive(s) can be used in an amount of less than 10 parts by weight, relative to 100 parts by weight of the polyamide.

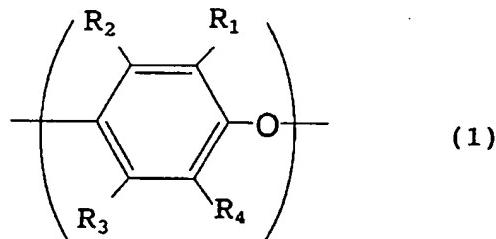
[0028]

<Polyphenylene ether>

Examples of polyphenylene ethers which can be used in the present invention include a homopolymer and a copolymer, each independently comprising a structural unit represented by the following formula (1):

[0029]

[Chemical formula 1]



[0030]

wherein O represents an oxygen atom, and each of R<sub>1</sub> to R<sub>4</sub> independently represents a hydrogen atom, a halogen atom, a primary or secondary lower alkyl group, a phenyl group, a haloalkyl group, an aminoalkyl group, a hydrocarbyloxy group or a halo hydrocar-

byloxy group (in which at least two carbon atoms are present between the halogen atom and the oxygen atom).

[0031]

Specific examples of polyphenylene ethers used in the present invention include poly(2,6-dimethyl-1,4-phenylene ether), poly(2-methyl-6-ethyl-1,4-phenylene ether), poly(2-methyl-6-phenyl-1,4-phenylene ether) and poly(2,6-dichloro-1,4-phenylene ether). Further examples of polyphenylene ethers include a copolymer of 2,6-dimethylphenol and another phenol (for example, a copolymer of 2,6-dimethylphenol and 2,3,6-trimethylphenol, and a copolymer of 2,6-dimethylphenol and 2-methyl-6-butylphenol, which are described in Examined Japanese Patent Application Publication No. Sho 52-17880).

[0032]

Among the above-mentioned polyphenylene ethers, preferred are poly(2,6-dimethyl-1,4-phenylene ether), a copolymer of 2,6-dimethylphenol and 2,3,6-trimethylphenol, and a mixture thereof.

[0033]

With respect to the methods for producing the polyphenylene ether used in the present invention, there is no particular limitation, and any conventional methods can be used. For example, there can be mentioned methods as described in U.S. Patent Nos. 3,306,874, 3,306,875, 3,257,357 and 3,257,358 and

Unexamined Japanese Patent Application Laid-Open Specification  
Nos. Sho 50-51197 and Sho 63-152628.

[0034]

With respect to the polyphenylene ether which can be used in the present invention, the reduced viscosity ( $\eta_{sp}/c$ ) thereof is preferably in the range of from 0.15 to 0.70 dl/g, more preferably from 0.20 to 0.60 dl/g, most preferably from 0.40 to 0.55 dl/g, as measured at 30 °C with respect to a 0.5 g/dl chloroform solution of the polyphenylene ether.

[0035]

In the present invention, a mixture of two or more different types of polyphenylene ethers having different reduced viscosities can be used without causing any problems. As examples of such a mixture, there can be mentioned a mixture of a polyphenylene ether having a reduced viscosity of 0.45 dl/g or less and a polyphenylene ether having a reduced viscosity of 0.50 dl/g or more, and a mixture of a low molecular weight polyphenylene ether having a reduced viscosity of 0.40 dl/g or less and a polyphenylene ether having a reduced viscosity of 0.50 dl/g or more, but the polyphenylene ether mixtures are not limited to those which are exemplified above.

[0036]

The polyphenylene ether used in the present invention may contain an organic solvent in an amount which is less than 5 parts by weight, relative to 100 parts by weight of the poly-

phenylene ether, wherein the organic solvent is the residual polymerization solvent used for producing the polyphenylene ether. It is difficult to remove the residual polymerization solvent completely by the drying operation performed after the polymerization reaction. The solvent usually remains in the polyphenylene ether in a concentration of several hundred ppm to several % by weight. The organic solvent mentioned herein, which is the residual polymerization solvent, may be at least one solvent selected from the group consisting of toluene, isomers of xylene, ethylbenzene, alcohols having 1 to 5 carbon atoms, chloroform, dichloromethane, chlorobenzene and dichlorobenzene.

[0037]

Further, the polyphenylene ether used in the present invention may be in a modified form or may be in the form of a mixture of an unmodified polyphenylene ether and a modified polyphenylene ether.

In the present invention, the "modified polyphenylene ether" means a polyphenylene ether which is modified with at least one modifier compound having at least one unsaturated bond selected from the group consisting of a carbon-carbon double bond and a carbon-carbon triple bond and having at least one group selected from the group consisting of a carboxylic acid group, an acid anhydride group, an amino group, a hydroxyl group and a glycidyl group.

[0038]

As examples of methods for producing the modified polyphenylene ether, there can be mentioned (1) a method in which polyphenylene ether is reacted with a modifier compound in either the presence or absence of a radical initiator at a reaction temperature which is 100 °C or higher and lower than the glass transition temperature of the polyphenylene ether, so that the reaction proceeds without causing the melting of the polyphenylene ether; (2) a method in which a polyphenylene ether and a modifier compound are melt-kneaded in either the presence or absence of a radical initiator at a temperature which is equal to or higher than the glass transition temperature of the polyphenylene ether and not higher than 360 °C, to thereby perform a reaction; and (3) a method in which a polyphenylene ether and a modifier compound are reacted in a solvent therefor in either the presence or absence of a radical initiator at a temperature which is lower than the glass transition temperature of the polyphenylene ether.

In the present invention, any of the above-mentioned methods (1) to (3) can be employed, but methods (1) and (2) are preferred.

[0039]

Next, a specific explanation is made on the above-mentioned modifier compound having at least one unsaturated bond selected from the group consisting of a carbon-carbon dou-

ble bond and a carbon-carbon triple bond and having at least one group selected from the group consisting of a carboxylic acid group, an acid anhydride group, an amino group, a hydroxyl group and a glycidyl group.

As examples of modifier compounds having a carbon-carbon double bond and a carboxylic acid group and/or an acid anhydride group, there can be mentioned maleic acid, fumaric acid, chloromaleic acid, cis-4-cyclohexene-1,2-dicarboxylic acid and acid anhydrides thereof. Among the above-mentioned compounds, fumaric acid, maleic acid and maleic anhydride are preferred, and fumaric acid and maleic anhydride are more preferred.

[0040]

Further, it is also possible to use a compound which is obtained by esterifying one or two carboxyl groups of any of the above-mentioned unsaturated dicarboxylic acids.

As examples of modifier compounds having a carbon-carbon double bond and a glycidyl group, there can be mentioned allyl-glycidyl ether, glycidyl acrylate, glycidyl methacrylate and epoxidized natural oils and fats.

Among the above-mentioned compounds, glycidyl acrylate and glycidyl methacrylate are especially preferred.

[0041]

As examples of modifier compounds having a carbon-carbon double bond and a hydroxyl group, there can be mentioned unsaturated alcohols represented by the following formula:

$C_nH_{2n-3}OH$  (wherein n is a positive integer), such as an allyl alcohol, 4-pentene-1-ol and 1,4-pentadiene-3-ol; and unsaturated alcohols represented by the following formulae:  $C_nH_{2n-5}OH$  and  $C_nH_{2n-7}OH$  (wherein n is a positive integer).

The above-mentioned modifier compounds may be used individually or in combination.

[0042]

The amount of the modifier compound used for producing the modified polyphenylene ether is preferably 0.1 to 10 parts by weight, more preferably 0.3 to 5 parts by weight, relative to 100 parts by weight of the polyphenylene ether.

When a modified polyphenylene ether is produced by using a radical initiator, the radical initiator is preferably used in an amount of 0.001 to 1 part by weight, relative to 100 parts by weight of the polyphenylene ether.

It is preferred that the amount of the modifier compound incorporated into the modified polyphenylene ether is 0.01 to 5 % by weight, more preferably 0.1 to 3 % by weight, based on the weight of the modified polyphenylene ether.

[0043]

The modified polyphenylene ether may contain unreacted modifier compound and/or a polymer of the modifier compound.

For reducing the amount of the unreacted modifier compound and/or the polymer of the modifier compound contained in the modified polyphenylene ether, if desired, a compound having an

amide linkage and/or an amino group may be added during the production of the modified polyphenylene ether.

[0044]

In the present invention, the "compound having an amide linkage" means a compound having a structure represented by the formula:  $-\text{NH}-\text{C}(=\text{O})-$ , and the "compound having an amino group" means a compound having a terminal  $-\text{NH}_2$  group. Specific examples of compounds having an amide linkage and/or an amino group include aliphatic amines, such as octylamine, nonylamine, tetramethylenediamine and hexamethylenediamine; aromatic amines, such as aniline, m-phenylenediamine, p-phenylenediamine, m-xylylenediamine and p-xylylenediamine; products obtained by reacting any of the above-mentioned amines with a carboxylic acid or a dicarboxylic acid; lactams, such as  $\epsilon$ -caprolactam; and polyamide resins, but the compounds having an amide linkage and/or an amino group are not limited to the compounds exemplified above.

[0045]

When the compound having an amide linkage and/or an amino group is used, it is preferred that the amount of the compound is 0.001 part by weight or more and less than 5 parts by weight, more preferably 0.01 part by weight or more and less than 1 part by weight, still more preferably 0.01 part by weight or more and less than 0.1 part by weight, relative to 100 parts by weight of the polyphenylene ether.

[0046]

Next, an explanation is made on the conductive carbon black used in the present invention.

In the present invention, it is preferred that the conductive carbon black has a dibutyl phthalate (DBP) oil absorption of at least 250 ml, more advantageously at least 300 ml, most advantageously at least 350 ml, per 100 g of the carbon black. In the present invention, the DBP oil absorption is a value obtained in accordance with ASTM D2414.

[0047]

Further, it is preferred that the conductive carbon black used in the present invention is a carbon black having a BET surface area of at least 200 cm<sup>2</sup>, more advantageously at least 400 cm<sup>2</sup>, per gram of the carbon black. Examples of such conductive carbon blacks which are commercially available include Ketjen black EC and a Ketjen black EC-600JD, both manufactured by Ketjen Black International Co.

[0048]

The amount of the conductive carbon black contained in the conductive resin composition is preferably in the range of from 0.2 to 5 parts by weight, relative to 100 parts by weight of the total of the components of the conductive resin composition exclusive of the conductive carbon black. Further, it is more preferred that the amount of the conductive carbon black is in the range of from 0.2 to 3 parts by weight, since the resin

composition has an excellent balance of impact resistance, melt-fluidity and conductivity.

[0049]

The conductive masterbatch of the present invention can be obtained by melt-kneading a polyamide and conductive carbon black.

The conductive masterbatch of the present invention can be in the form of pellets, a powder or granules, and it is preferred that the masterbatch is in the form of pellets, more advantageously pellets having a diameter of 1.5 to 3.5 mm and a length of 2.0 to 3.5 mm. Hereinbelow, the pellets of the conductive masterbatch are referred to as "master pellets".

[0050]

The conductive resin composition of the present invention can be obtained by melt-kneading a polyamide, a polyphenylene ether and the conductive masterbatch.

The amount of the conductive carbon black contained in the conductive masterbatch is preferably 5 to 40 % by weight, more preferably 5 to 25 % by weight, still more preferably 6 to 15 % by weight, most preferably 6 to 10 % by weight, based on the total weight of the components of the masterbatch. Especially when the amount of the conductive carbon black in the conductive masterbatch is 6 to 10 % by weight, not only can the lowering of the molecular weight of the polyamide contained in the masterbatch be prevented, but also the productivity of the mas-

terbatch can be improved.

[0051]

In the present invention, it is important that a part of the conductive carbon black in the masterbatch is present in the form of at least one agglomerated particle. When a part of the conductive carbon black in the masterbatch is present in the form of agglomerated particle(s), the conductivity of the masterbatch is improved as compared to that of a masterbatch in which the conductive carbon black is not present in the form of agglomerated particle(s). In the present invention, it is preferred that an agglomerated particle of the conductive carbon black has a major axis of 20 to 100  $\mu\text{m}$ . The number of the agglomerated particle(s) having a major axis of 20 to 100  $\mu\text{m}$  or more is preferably 1 to 100, more preferably 1 to 50, still more preferably 2 to 40, most preferably 2 to 30, as observed under an optical microscope with respect to a contiguous area of 3  $\text{mm}^2$ .

[0052]

In the present invention, the size and number of the agglomerated particle(s) of the conductive carbon black are determined by a method which is explained below. Specifically, the masterbatch is cut by means of a microtome equipped with a glass knife to thereby obtain cross-section which has a mirror surface. The light reflected from the cross-section is observed with a magnification of  $\times 50$  under an optical microscope

(for example, "PME3" manufactured and sold by Olympus Optical Co. Ltd.), and a photomicrograph of cross-section is taken. With respect to a contiguous area of 3 mm<sup>2</sup> in the photomicrograph, the number of agglomerated particles (each independently having a major axis of 20 to 100 µm) is counted by visual observation. When the master pellet has a cylindrical or rectangular parallelepiped shape, the pellet is cut in a direction substantially perpendicular to the longitudinal direction of the pellet, thereby obtaining a cross-section, and the cross-section is observed (see Fig. 1).

[0053]

When the master pellet has a granular or spherical shape, the pellet is cut along a plane including the center of the pellet, and the resultant cross-section of the pellet is observed (see Fig. 2). The number of the agglomerated particles is counted with respect to at least three cross-sections which are, respectively, obtained from different pellets, and an average value of the measured values is calculated. When a cross-section having a contiguous area of 3 mm<sup>2</sup> or more cannot be obtained, a plurality of cross-sections are observed to count the number of the agglomerated particles present in the cross-sections having a total area of 3 mm<sup>2</sup>.

[0054]

In the present invention, it is preferred that the central line average roughness (Ra) of each of the master pellets is in

the range of from 0.3 to 2.0  $\mu\text{m}$ , more advantageously from 0.4 to 1.5  $\mu\text{m}$ , in terms of an average value of the surface roughness (Ra) values as measured by a surface roughness gauge with respect to a plurality of surface portions of each of the pellets.

The above-mentioned surface roughness gauge is an apparatus for measuring the unevenness of a surface, and the surface roughness gauge also includes a scanning probe microscope.

[0055]

In the present invention, when the master pellet has a flat portion, the central line average roughness (Ra) of the flat portion is measured using a surface roughness gauge Surfcom 579A (manufactured and sold by Tokyo Seimitsu Co., Ltd.) wherein the measuring length is 2.5 mm. When the master pellet does not have a flat portion at which the measuring length of 2.5 mm can be obtained, the measurement is conducted with respect to a plurality of flat portions so as to obtain a total measuring length of 2.5 mm. In the case of a master pellet (such as a granular master pellet) having no flat portion, an image ( $80 \mu\text{m} \times 80 \mu\text{m}$ ) of the surface of the pellet is observed by using a scanning probe microscope (SPA300HV, manufactured and sold by Seiko Instruments Inc.), followed by three dimensional correction of the image to obtain an image showing the surface profile of the pellet. Using the obtained image showing the surface profile of the pellet, the central line average

roughness ( $R_a$ ) is obtained. In the present invention, the surface roughness of the pellet is an average value of 10 ( $R_a$ ) values.

[0056]

In the present invention, the surface roughness (central line average roughness:  $R_a$ ) value has a relationship to the luster of the pellet and the state of dispersion of the conductive carbon black in the pellet. That is, when substantially no agglomerated particle is present in the pellet, the luster of the pellet becomes high and the surface roughness becomes small. On the other hand, when the agglomerated particles are present in the pellet, the luster of the pellet becomes low and the surface roughness becomes large.

[0057]

As a preferred method for producing the conductive masterbatch, there can be mentioned a method in which the raw materials for the masterbatch are melt-kneaded by using a twin-screw extruder or a kneader. Especially preferred is a method in which a polyamide is melted, followed by addition of a conductive carbon black. As a specific example of such a method, there can be mentioned a method which uses a twin-screw extruder or kneader having at least one first inlet and at least one second inlet which are, respectively, provided at an upstream portion and a downstream portion of the extruder or kneader, wherein, in this method, a polyamide is fed from the

first inlet of the extruder or kneader to thereby melt the polyamide, and a conductive carbon black is added to the molten polyamide in the extruder or kneader from the second inlet thereof, followed by melt-kneading of the resultant mixture.

[0058]

The conductive resin composition of the present invention may contain a styrene-containing thermoplastic resin in an amount of less than 50 parts by weight, relative to 100 parts by weight of the total of the polyamide and the polyphenylene ether. As examples of styrene-containing thermoplastic resins used in the present invention, there can be mentioned a polystyrene (homopolymer), a rubber-modified polystyrene (HIPS), a styrene-acrylonitrile copolymer (AS resin) and a styrene-rubber polymer-acrylonitrile copolymer (ABS resin).

Any of conventional additives which can be used for a polyphenylene ether can be added to the conductive resin composition in an amount of less than 10 parts by weight, relative to 100 parts by weight of the polyphenylene ether.

[0059]

Further, the conductive resin composition of the present invention may contain an impact modifier.

As the impact modifier added to the conductive resin composition of the present invention, for example, it is possible to use at least one polymer selected from the group consisting of an aromatic vinyl compound/conjugated diene block copolymer

comprising a polymer block composed mainly of aromatic vinyl monomer units and a polymer block composed mainly of conjugated diene monomer units and a hydrogenation product thereof, and an ethylene/α-olefin copolymer.

[0060]

Specific examples of aromatic vinyl compounds used for producing the aromatic vinyl compound/conjugated diene block copolymer used in the present invention include styrene, α-methyl styrene and vinyl toluene. These compounds can be used individually or in combination. Among the above-exemplified compounds, styrene is especially preferred.

Specific examples of conjugated dienes used for producing the aromatic vinyl compound/conjugated diene block copolymer used in the present invention include butadiene, isoprene, piperylene and 1,3-pentadiene. These compounds can be used individually or in combination. Among the above-exemplified compounds, preferred are butadiene, isoprene and a mixture thereof.

[0061]

With respect to the microstructure of a soft segment (composed of the conjugated diene monomer units) of the above-mentioned block copolymer, it is preferred that the 1,2-vinyl bond content or the total content of the 1,2-vinyl bond and the 3,4-vinyl bond is 5 to 80 %, more advantageously 10 to 50 %, most advantageously 10 to 40 %.

[0062]

It is preferred that the block copolymer used in the present invention has a block configuration selected from the group consisting of A-B, A-B-A and A-B-A-B, wherein A represents an aromatic vinyl polymer block and B represents a conjugated diene polymer block. The block copolymer used in the present invention can be a mixture of block copolymers having different block configurations.

Among the above-mentioned block configurations, A-B-A and A-B-A-B are preferred. The block copolymer can be a mixture of different block copolymers having the above-mentioned block configurations.

[0063]

Further, it is preferred that the aromatic vinyl compound/conjugated diene block copolymer used for the conductive resin composition of the present invention is a hydrogenated block copolymer. The "hydrogenated block copolymer" herein means a copolymer which is obtained by hydrogenating any of the above-mentioned aromatic vinyl compound/conjugated diene block copolymers wherein the degree of hydrogenation of the aliphatic double bonds in the conjugated diene polymer block is more than 0 % and up to 100 %. The degree of hydrogenation of the hydrogenated block copolymer is preferably 50 % or more, more preferably 80 % or more, most preferably 98 % or more.

[0064]

In the present invention, a mixture of an unhydrogenated

block copolymer and a hydrogenated block copolymer can be used without causing any problem.

With respect to the block copolymer used in the conductive resin composition of the present invention, it is preferred that the block copolymer is a mixture of a low molecular weight block copolymer and a high molecular weight block copolymer. Specifically, it is preferred to use a mixture of a low molecular weight block copolymer having a number average molecular weight of less than 120,000 and a high molecular weight block copolymer having a number average molecular weight of 120,000 or more. It is more preferred to use a mixture of a low molecular weight block copolymer having a number average molecular weight of less than 100,000 and a high molecular weight block copolymer having a number average molecular weight of 200,000 or more.

[0065]

In the present invention, the number average molecular weight is measured by means of a gel permeation chromatography (GPC) apparatus (for example, GPC SYSTEM 21, manufactured and sold by Showa Denko Co.), using an ultraviolet spectrometric detector (for example, UV-41, manufactured and sold by Showa Denko Co.) and a calibration curve obtained with respect to standard polystyrene samples. The conditions employed for measuring the number average molecular weight are as follows.

[0066]

(Conditions)

Solvent: chloroform,

Temperature: 40 °C,

Columns: columns for the sample (K-G, K-800RL and K-800R)

and columns for the reference (K-805L, 2 columns),

Flow rate: 10 ml/min,

Wavelength used for detection: 254 nm, and

Pressure: 15 to 17 kg/cm<sup>2</sup>.

[0067]

In the measurement of the number average molecular weight, a low molecular weight component by-produced due to the deactivation of a polymerization catalyst may be detected, but such a low molecular weight component is ignored in the calculation of the molecular weight. In general, a correctly calculated molecular weight distribution (weight average molecular weight/number average molecular weight ratio) is in the range of from 1.0 to 1.2.

The weight ratio of the low molecular weight block copolymer to the high molecular weight block copolymer (low molecular weight block copolymer/high molecular weight block copolymer weight ratio) in the mixture of the low molecular weight block copolymer and the high molecular weight block copolymer is generally in the range of from 95/5 to 5/95, preferably 90/10 to 10/90.

[0068]

Further, in the present invention, when the low molecular weight block copolymer used in the above-mentioned mixture comprises an aromatic vinyl polymer block having a number average molecular weight of 20,000 or more, it becomes possible to improve the heat resistance of the conductive resin composition in addition to the impact resistance.

The number average molecular weight of an aromatic vinyl polymer block of a block copolymer can be calculated from the number average molecular weight of the block copolymer mentioned above in accordance with the following formula:

[0069]

$$Mn(a) = \{Mn \times a/(a + b)\}/N$$

wherein  $Mn(a)$  represents the number average molecular weight of the aromatic vinyl polymer block;  $Mn$  represents the number average molecular weight of the block copolymer; "a" represents the % by weight of the total of the aromatic vinyl polymer blocks, based on the weight of the block copolymer; "b" represents the % by weight of the total of the conjugated diene polymer blocks, based on the weight of the block copolymer; and  $N$  represents the number of the aromatic vinyl polymer blocks in the block copolymer.

[0070]

In the present invention, it is preferred that the aromatic vinyl polymer block content of the low molecular weight

block copolymer is 55 % by weight or more and less than 90 % by weight. When the aromatic vinyl polymer block content of the low molecular weight block copolymer is within the above-mentioned range, the heat resistance of the conductive resin composition can be improved, so that such a resin composition can be very advantageously used.

[0071]

Further, in the present invention, when the low molecular weight block copolymer is a mixture of a block copolymer having an aromatic vinyl polymer block content of 55 % by weight or more and less than 90 % by weight and a block copolymer having an aromatic vinyl polymer block content of 20 % by weight or more and less than 55 % by weight, it becomes possible to improve the melt-fluidity of the conductive resin composition.

[0072]

Before the block copolymer is mixed with the conductive resin composition of the present invention, the block copolymer may be mixed with an oil composed mainly of a paraffin. The addition of an oil composed mainly of paraffin to the block copolymer leads to the improvement in the processability of the resin composition. The amount of the oil contained in the block copolymer is preferably 1 to 70 parts by weight, relative to 100 parts by weight of the block copolymer. When the block copolymer contains more than 70 parts by weight of the oil, the handling properties of the block copolymer become poor.

[0073]

In the present invention, the oil composed mainly of a paraffin means a mixture of hydrocarbon compounds each independently having a weight average molecular weight of from 500 to 10,000, which mixture comprises an aromatic ring-containing compound, a naphthenic ring-containing compound and a paraffin compound, wherein the content of the paraffin compound is 50 % by weight or more. It is preferred that the oil contains 50 to 90 % by weight of a paraffin compound, 10 to 40 % by weight of a naphthenic ring-containing compound and not more than 5 % by weight of an aromatic ring-containing compound, based on the weight of the oil.

[0074]

Such an oil composed mainly of a paraffin is commercially available. For example, there can be mentioned PW 380 which is manufactured and sold by Idemitsu Kosan Co., Ltd.

The above-mentioned aromatic vinyl compound/conjugated diene block copolymer may be a mixture of different block copolymers so long as each of the block copolymers does not adversely affect the properties of the resin composition of the present invention. For example, the block copolymer may be a mixture of block copolymers having different block configurations, a mixture of block copolymers containing different aromatic vinyl monomer units, a mixture of block copolymers containing different conjugated diene monomer units, a mixture of block copoly-

mers having different 1,2-vinyl contents or different total contents of 1,2-vinyl bond and 3,4-vinyl bond, a mixture of block copolymers having different aromatic vinyl monomer unit contents, and a mixture of block copolymers having different degrees of hydrogenation.

[0075]

As a specific example of ethylene- $\alpha$ -olefin copolymers which can be used in the present invention, there can be mentioned one which is described in Unexamined Japanese Patent Application Laid-Open Specification No. 2001-302911.

Further, the impact modifier used in the conductive resin composition of the present invention may be a modified impact modifier or a mixture of a modified impact modifier and an unmodified impact modifier.

The modified impact modifier mentioned herein means an impact modifier which is modified with at least one modifier compound having at least one unsaturated bond selected from the group consisting of a carbon-carbon double bond and a carbon-carbon triple bond and having at least one group selected from the group consisting of a carboxylic acid group, an acid anhydride group, an amino group, a hydroxyl group and a glycidyl group.

[0076]

As examples of methods for producing the modified impact modifier, there can be mentioned (1) a method in which an im-

pact modifier and a modifier compound are melt-kneaded in either the presence or absence of a radical initiator at a reaction temperature which is not lower than the softening temperature of the impact modifier and not higher than 250 °C, to thereby perform a reaction; (2) a method in which an impact modifier and a modifier compound are reacted with each other in a solvent therefor in either the presence or absence of a radical initiator at a temperature which is not higher than the softening temperature of the impact modifier; and (3) a method in which an impact modifier is reacted with a modifier compound in either the presence or absence of a radical initiator at a reaction temperature which is not higher than the softening temperature of the impact modifier, without causing the melting of the impact modifier. Any one of the above-mentioned methods (1) to (3) can be employed, but method (1) is preferred, and method (1) performed in the presence of a radical initiator is most preferred.

[0077]

As the above-mentioned modifier compound having at least one unsaturated bond selected from the group consisting of a carbon-carbon double bond and a carbon-carbon triple bond and having at least one group selected from the group consisting of a carboxylic acid group, an acid anhydride group, an amino group, a hydroxyl group and a glycidyl group, any of those which are exemplified above as the modifier compound used for

modifying a polyphenylene ether can be used.

[0078]

It is preferred that the conductive resin composition of the present invention contains 30 to 70 parts by weight of a polyamide, 20 to 50 parts by weight of a polyphenylene ether and 5 to 30 parts by weight of an impact modifier, relative to 100 parts by weight of the total of the polyamide, polyphenylene ether and impact modifier. It is more preferred that the conductive resin composition of the present invention contains 40 to 60 parts by weight of the polyamide, 30 to 40 parts by weight of the polyphenylene ether and 5 to 15 parts by weight of the impact modifier, relative to 100 parts by weight of the total of the polyamide, polyphenylene ether and impact modifier.

[0079]

Further, in the present invention, a compatibility agent can be incorporated into the conductive resin composition during the production thereof. In general, a compatibility agent is used mainly for the purpose of improving the physical properties of a polyamide-polyphenylene ether alloy. The compatibility agent which can be used in the present invention is a multifunctional compound which interacts with one or both of the polyphenylene ether and the polyamide.

In the conductive resin composition of the present invention, it is preferred that the compatibility of the polyamide

with the polyphenylene ether is improved by using the compatibility agent.

[0080]

Examples of compatibility agents which can be used in the conductive resin composition of the present invention include those which are described in Unexamined Japanese Patent Application Laid-Open Specification Nos. Hei 8-48869 and Hei 9-124926. All of the conventional compatibility agents described in these patent documents can be used in the present invention, and the compatibility agents can be used individually or in combination.

Among various conventional compatibility agents, especially preferred are maleic acid, maleic anhydride and citric acid.

[0081]

The amount of the compatibility agent used in the conductive resin composition of the present invention is preferably 0.1 to 20 parts by weight, more preferably 0.1 to 10 parts by weight, relative to 100 parts by weight of the total of the polyamide and the polyphenylene ether which are contained in the resin composition of the present invention.

In the present invention, in addition to the above-mentioned components of the conductive resin composition, if desired, an additional component can be added to the conductive resin composition so long as the additional component does

not adversely affect the excellent properties of the resin composition of the present invention.

[0082]

The additional components used in the present invention are explained below.

Examples of additional components include thermoplastic resins, such as a polyester and a polyolefin; inorganic fillers (such as talc, kaolin, xonotlite, wollastonite, titanium oxide, potassium titanate and a glass fiber); conventional adhesion modifiers which enhance the affinity between an inorganic filler and a resin; flame retardants (such as a halogenated resin, a silicone flame retardant, magnesium hydroxide, aluminum hydroxide, an organic phosphoric ester compound, ammonium polyphosphate and red phosphorus), fluororesins having an effect to prevent the dripping of flaming particles; plasticizers (such as an oil, a low molecular weight polyolefin, a polyethylene glycol and a fatty ester); auxiliary flame retardants, such as antimony trioxide; carbon black as a pigment; conductivity imparting agents, such as a carbon fiber, a carbon nanotube and a carbon nanofiber; antistatic agents; various peroxides; zinc oxide; zinc sulfide; antioxidants; ultraviolet absorbers; and light stabilizers.

In the present invention, the amount of the additional component added to the resin composition is not more than 100 parts by weight, relative to 100 parts by weight of the total

of the polyamide, the polyphenylene ether and the impact modifier.

[0083]

As specific examples of processing apparatuses which can be used to prepare the conductive resin composition of the present invention, there can be mentioned a single-screw extruder, a twin-screw extruder, a roll, a kneader, a Brabender Plastograph and a Banbury mixer. Among these apparatuses, preferred is a twin-screw extruder, and especially preferred is a twin-screw extruder provided with a first inlet and at least one second inlet which are, respectively, formed at an upstream portion and a downstream portion of the extruder.

With respect to the melt-kneading temperature used for producing the resin composition, there is no particular limitation. In general, an appropriate temperature for obtaining a desired resin composition is selected from the range of from 240 to 360 °C.

[0084]

As preferred examples of the method for producing the conductive resin composition of the present invention, there can be mentioned a method which uses a twin-screw extruder having a first inlet and a second inlet which are formed, respectively, at an upstream portion and downstream portion of the extruder, wherein an impact modifier and a polyphenylene ether are fed to the extruder from the first inlet, thereby melt-kneading to-

gether the impact modifier and the polyphenylene ether at the upstream portion of the extruder, followed by feeding a conductive carbon black to the extruder from the second inlet, thereby melt-kneading together the impact modifier, the polyphenylene ether, the polyamide and the conductive carbon black at the downstream portion of the extruder, wherein the method comprises the steps: (1) providing a conductive masterbatch comprising a polyamide and a conductive carbon black, wherein a part of the conductive carbon black is present in the form of an agglomerated particle having a major axis of 20 to 100  $\mu\text{m}$ , and (2) adding the conductive masterbatch to a molten polyphenylene ether. More preferred is a method in which the conductive masterbatch and a polyamide are simultaneously added to the molten polyphenylene ether.

[0085]

The thus obtained conductive resin composition can be molded into various molded articles by various conventional methods, such as injection molding.

Examples of molded articles include parts for electrical or electronic appliances, such as an IC tray, a chassis and cabinet of various disc players; parts for office automation machines, such as various computers and peripheral equipment therefor; and mechanical parts; a cowl for a motorcycle; exterior parts for automobiles, such as a fender, a door panel, a front panel, a rear panel, a locker panel, a rear bumper panel,

a back door garnish, an emblem garnish, a panel for a feeding port of a fuel, an over fender, an outer door handle, a door mirror housing, a bonnet air intake, a bumper, a bumper guard, a roof rail, a roof rail leg, a pillar, a pillar cover, a wheel cover, various aero parts (such as a spoiler), various moles and emblems for an automobile; and interior parts for automobiles, such as an instrument panel, a console box and a trim.

[0086]

Among the above-exemplified molded articles, the conductive resin composition of the present invention is suitable for producing an automobile outer panel, especially a fender of an automobile, which can be electrostatically coated.

The surface roughness measured with respect to an automobile fender in the present invention is preferably in the range of from 0.05 to 1  $\mu\text{m}$ , from the viewpoint of appearance of the outer panel after the electrostatic coating.

[0087]

In the present invention, the surface roughness of an automobile fender can be determined as follows. From the automobile fender, eight different surface portions are selected. With respect to each of the selected eight portions, an area of  $20 \mu\text{m} \times 20 \mu\text{m}$  is observed under a scanning probe microscope (SPA300HV, manufactured and sold by Seiko Instruments Inc.). The obtained images of the eight portions are three dimensionally corrected to obtain images showing the surface profiles of

the eight portions. Using the resultant three-dimensionally corrected images, the difference between the maximum height and the minimum height at each of the eight surface portions is determined, followed by the calculation of the average value of the obtained eight difference values.

[0088]

[Examples]

Hereinbelow, the present invention will be described in more detail with reference to the following Examples and Comparative Examples, which should not be construed as limiting the scope of the present invention.

[0089]

(Raw Materials)

In the following Examples and Comparative Examples, the following raw materials were used.

(1) Polyamide (hereinafter, abbreviated to "PA")

(1-1) Polyamide 6 (hereinafter, abbreviated to "PA6")

Trade name: UBE Nylon 6 SF1013A (manufactured and sold by UBE INDUSTRIES, LTD.)

(1-2) Polyamide 66 (hereinafter, abbreviated to "PA66")

Number average molecular weight = 14,000

Concentration of terminal amino groups =

30 milliequivalent/kg

Concentration of terminal carboxyl groups =

100 milliequivalent/kg

[0090]

- (2) Conductive carbon black (hereinafter, abbreviated to "KB")

Trade name: Ketjenblack EC-600JD (manufactured and sold by Ketjenblack International Corporation)

[0091]

- (3) Polyphenylene ether (hereinafter, abbreviated to "PPE")  
Poly(2,6-dimethyl-1,4-phenylene ether) (manufactured and sold by Asahi Kasei Kabushiki Kaisha)

- (3-1) Polyphenylene ether (hereinafter, abbreviated to "PPE-1")

Reduced viscosity: 0.52 dl/g (measured at 30 °C using a 0.5 g/dl chloroform solution of PPE-1)

- (3-2) Polyphenylene ether (hereinafter, abbreviated to "PPE-2")

Reduced viscosity: 0.42 dl/g (measured at 30 °C using a 0.5 g/dl chloroform solution of PPE-2)

- (3-3) Polyphenylene ether modified with maleic anhydride (hereinafter, abbreviated to "M-PPE")

M-PPE was prepared by adding 0.1 part by weight of a radical initiator and 1.5 parts by weight of maleic anhydride to 100 parts by weight of PPE-2, followed by melt-kneading in a twin-screw extruder at a cylinder temperature of 320 °C.

The amount of the maleic anhydride incorporated into

the modified polyphenylene ether was 0.5 % by weight, based on the weight of the modified polyphenylene ether.

[0092]

(4) Polystyrene (hereinafter, abbreviated to "PS")

Trade name: A & M Polystyrene 685 (manufactured and sold by A & M Styrene Co., Ltd.)

[0093]

(5) Block copolymer

(5-1) Polystyrene/hydrogenated polybutadiene/polystyrene block copolymer (hereinafter, abbreviated to "SEBS-1")

Number average molecular weight = 246,000

Total styrene content = 33 %

(5-2) Polystyrene/hydrogenated polybutadiene/polystyrene block copolymer (hereinafter, abbreviated to "SEBS-2")

Number average molecular weight = 77,000

Total styrene content = 67 %

[0094]

(6) Compatibility agent

(6-1) Citric acid (manufactured and sold by Wako Pure Chemical Industries, Ltd.)

(6-2) Maleic anhydride (manufactured and sold by Mitsubishi Chemical Corporation)

[0095]

(Measuring methods)

The methods for measuring the number of agglomerated particles of conductive carbon black (hereinafter, referred to as "number of agglomerated particles"), surface roughness, impact resistance, heat resistance and conductivity (surface resistivity and volume resistivity) are explained below.

[0096]

<Number of agglomerated particles>

A pellet of masterbatch was cut by means of a microtome equipped with a glass knife to thereby obtain a cross-section which has a mirror surface. The light reflected from the cross-section was observed with a magnification of  $\times 50$  under an optical microscope ("PME3", manufactured and sold by Olympus Optical Co. Ltd.), and a photomicrograph of the cross-section was taken. With respect to a contiguous area of  $3 \text{ mm}^2$  in the photomicrograph, the number of agglomerated particles each independently having a major axis of 20 to  $100 \mu\text{m}$  was counted by visual observation. Since the master pellet had a cylindrical shape, the pellet was cut in a direction substantially perpendicular to the longitudinal direction of the pellet, thereby obtaining a cross-section, and the resultant cross-section was observed. Three cross-sections which were, respectively, obtained from different pellets were observed. The number of agglomerated particles was determined in terms of the average

value of the three counted values at three cross-sections.

[0097]

<Surface roughness>

The central line average roughness (Ra) of a master pellet was measured by using a surface roughness gauge (Surfcom 579 A, manufactured and sold by Tokyo Seimitsu Co., Ltd.), wherein the Ra value was measured with respect to a flat portion of the pellet. The measurement was performed under conditions wherein the moving rate of the contact finger was 0.3 mm/sec and the measuring length was 2.5 mm. The measurement was conducted at ten flat portions of the pellet, and an average value of the ten measured values was calculated.

[0098]

<Izod impact strength and deflection temperature under load>

Pellets of a resin composition were molded by using a molding machine (TOSHIBA IS-80EPN) under conditions wherein the cylinder temperature was 280 °C and the mold temperature was 80 °C, to thereby obtain test strips having a thickness of 3.2 mm and a thickness of 6.4 mm, respectively. The notched Izod impact strength of the 3.2 mm-thick test strip was measured in accordance with ASTM D256. Further, the deflection temperature under load (HDT) of the 6.4 mm-thick test strip was measured in accordance with ASTM D648 under a load of 1.82 MPa.

[0099]

<Surface resistivity and volume resistivity-1>

Pellets of a resin composition were molded into a test plate (size: 100 × 50 × 2.5 mm) by using a molding machine (TOSHIBA IS-80EPN) under conditions wherein the cylinder temperature was 280 °C and the mold temperature was 80 °C. The surface resistivity and the volume resistivity of the obtained test plate were measured by using a high resistivity meter (MCP-HT450, manufactured and sold by MITSUBISHI CHEMICAL CORPORATION) at 500 V. The measurement was performed with respect to five different test plates (each prepared in the above-mentioned manner) and each of the surface resistivity value and the volume resistivity value was obtained in terms of the average value of the five measured values.

[0100]

<Volume resistivity-2>

In accordance with the method described in the Examples of the patent document 3, pellets of a resin composition were molded into a dumbbell-shaped bar described under ISO 294 by using a molding machine (Toshiba IS-80EPN) under conditions wherein the cylinder temperature was 280 °C and the mold temperature was 80 °C. The thus obtained dumbbell-shaped bar was used as a test specimen. Both ends of the test specimen were broken off so as to obtain a portion with a uniform cross section of 10 mm × 4 mm and a length of about 70 mm with a fractured surface at both ends. A silver-containing coating composition was applied to the broken ends of the obtained portion

and the volume resistivity between the silver-coated ends was measured by using a digital insulation testing apparatus (DG 525, SANWA ELECTRIC INSTRUMENT CO., LTD.) wherein a voltage of 250 V was applied. The measurement was performed with respect to five different test specimens and the volume resistivity value was obtained in terms of the average value of the five measured values.

[0101]

<Surface roughness value of an automobile fender>

By using a scanning probe microscope (SPA300HV, manufactured and sold by Seiko Instruments Inc.), the surface roughness of an automobile fender was determined. Specifically, eight different surface portions (each having an area of  $20 \mu\text{m} \times 20 \mu\text{m}$ ) were arbitrarily selected from an automobile fender, and observed under the scanning probe microscope. The resultant images of the eight surface portions were subjected to a three-dimensional correction to obtain images showing the surface profiles of the eight surface portions, and the difference between the maximum height and the minimum height (surface roughness value) was measured with respect to each of the eight surface portions. The average value of the obtained eight difference values was defined as the surface roughness of the automobile fender. SI-DF20 (manufactured and sold by Seiko Instruments Inc.) was used as a cantilever of the microscope, and the observation was made under conditions wherein the atmos-

phere was air, the temperature was 25 °C, the scanning frequency was 0.5 Hz, the detection mode was dynamic force mode (DFM) and the vibration amplitude was 0.8 V.

[0102]

[Comparative Example 1]

By using a twin-screw extruder (ZSK-25, manufactured and sold by Krupp Werner & Pfleiderer GmbH, Germany) which had one inlet at an upstream portion thereof and another inlet at a downstream portion thereof, 92 parts by weight of polyamide 6 and 8 parts by weight of conductive carbon black were uniformly melt-kneaded at a cylinder temperature of 270 °C in accordance with the method described in the single Example of patent document 2, to thereby produce a masterbatch in the form of pellets (master pellets) having a surface luster. In the extruder used, the screws were so designed as to generate a high shearing force, such that the surface roughness Ra of the master pellets becomes less than 0.3 µm. With respect to the obtained master pellets (hereinafter, abbreviated to "PA/KB-MB1"), the number of agglomerated particles contained therein and the surface roughness Ra at the flat portions thereof were measured (see Fig. 3). Further, by using the obtained masterbatch, a resin composition was produced in accordance with the method described in the single Example of patent document 2. Specifically, pellets of a resin composition comprising polyamide 6, a polyphenylene ether and conductive carbon black were produced

at a cylinder temperature of 300 °C. The obtained pellets of the resin composition were molded into test specimens, and the surface resistivity and the volume resistivity-1 of the resin composition were measured using the test specimens. The properties of the resin composition are shown in Table 1, together with the formulation of the resin composition.

[0103]

[Example 1]

A conductive masterbatch comprising polyamide 6 and conductive carbon black (polyamide 6/conductive carbon black masterbatch) which was in the form of pellets (master pellets) having a low surface luster was produced in substantially the same manner as in Comparative Example 1, except that the design of the screws of the twin-screw extruder (ZSK-25, manufactured and sold by Krupp Werner & Pfleiderer GmbH, Germany) (used for melt-kneading 92 parts by weight of polyamide 6 and 8 parts by weight of conductive carbon black at a cylinder temperature of 270 °C) was changed. Specifically, the production of the masterbatch was conducted while controlling the melt-kneading conditions (i.e., the rotation rate of the screws, extrusion rate and the like) to obtain master pellets having a surface roughness Ra of 0.3 µm or more (the melt-kneading conditions employed in Example 1 were mild as compared to those employed in Comparative Example 1). With respect to the obtained master pellets (hereinafter, abbreviated to "PA/KB-MB2"), the number

of agglomerated particles contained therein and the surface roughness Ra of the flat portions thereof were measured (see Fig. 4). Further, by using the obtained conductive masterbatch, pellets of a resin composition comprising polyamide 6, a polyphenylene ether and conductive carbon black were produced in substantially the same manner as in Comparative Example 1. The obtained pellets of the resin composition were molded into test specimens, and the surface resistivity and the volume resistivity of the resin composition were measured using the test specimens. The properties of the resin composition are shown in Table 1, together with the formulation of the resin composition.

[0104]

[Table 1]

Feeding inlet of the ex- truder	Components of the resin composition	units	Comparative	Example 1
			Example 1	Formulation of resin
Top	PPE	Parts by weight	38	38
	Citric acid		1	1
	PS		12	12
	PA6		6.5	6.5
	PA/KB-MB1		37.5	-
	PA/KB-MB2		-	37.5
Results of measurement (masterbatch: PA6/KB-MB1 and PA6/KB-MB2)				
Number of agglomerated particles (major axis: 20 to 100 µm)		pieces	0	16
Number of agglomerated particles (major axis: 30 to 100 µm)		pieces	0	9
Number of agglomerated particles (major axis: more than 100 µm)		pieces	0	0
Surface roughness Ra		µm	0.22	1.04
Results of measurement (resin composition: PA6/PPE/KB)				
Surface resistivity		Ω	$4.5 \times 10^9$	$3.6 \times 10^7$
Volume resistivity-1		Ω · cm	$3.7 \times 10^{10}$	$3.0 \times 10^7$

[0105]

From Table 1, it is apparent that the conductivity of a resin composition comprising a polyamide, a polyphenylene ether and conductive carbon black can be remarkably improved by the use of a masterbatch in which the conductive carbon black is present in the form of at least one agglomerated particle.

[0106]

[Comparative Example 2]

In accordance with the method described in the single Ex-

ample of patent document 2, 90 parts by weight of polyamide 66 and 10 parts by weight of conductive carbon black were uniformly melt-kneaded at a cylinder temperature of 270 °C, thereby producing a conductive masterbatch in the form of pellets (master pellets) having a surface luster. In the extruder used, the screws were so designed as to generate a high shearing force, such that the surface roughness Ra of the resultant pellets becomes less than 0.3 µm. With respect to the obtained master pellets (hereinafter, abbreviated to "PA/KB-MB3"), the number of agglomerated particles contained therein and the surface roughness Ra of the flat portions thereof were measured. Further, using the obtained master pellets, a resin composition in the form of pellets was produced by using a twin-screw extruder (ZSK-25, manufactured and sold by Krupp Werner & Pfleiderer GmbH, Germany) having one inlet at an upstream portion thereof and another inlet at a downstream portion thereof. Specifically, the conductive masterbatch (PA/KB-MB3) were melt-kneaded together with a polyphenylene ether, a block copolymer and polyamide 66 in the extruder at a cylinder temperature of 300 °C, wherein the polyphenylene ether and the block copolymer were fed to the extruder from the inlet provided at an upstream portion thereof, while feeding polyamide 66 and the conductive masterbatch (PA/KB-MB3) to the extruder from the inlet provided at a downstream portion thereof, thereby obtaining a resin composition comprising polyamide 66, the polyphenylene ether and

the conductive carbon black. The obtained resin composition was molded into test specimens, and the volume resistivity-2, deflection temperature under load and Izod impact strength of the resin composition were evaluated using the test specimen. The results are shown in Table 2, together with the formulation of the resin composition.

[0107]

[Comparative Example 3]

A resin composition was produced using a twin-screw extruder (ZSK-25, manufactured and sold by Krupp Werner & Pfleiderer GmbH, Germany) having an inlet at an upstream portion thereof and two inlets at downstream portions thereof (with respect to the two inlets provided at downstream portions of the extruder, the first and second inlets, as view in the direction of from the upstream to downstream of the extruder, are referred to as "1st downstream-side inlet" and "2nd downstream-side inlet"). In accordance with the method described in patent document 3, a polyphenylene ether, a block copolymer, polyamide 66 and conductive carbon black were melt-kneaded together at a cylinder temperature of 300 °C, wherein the polyphenylene ether and the block copolymer were fed to the extruder from the inlet at an upstream portion thereof and the polyamide 66 was fed to the extruder from the 1st downstream-side inlet, thereby compatibilizing the polyphenylene ether with the polyamide, while feeding the conductive carbon black from the 2nd down-

stream-side inlet, thereby producing a resin composition in the form of pellets, which composition comprises polyamide 66, a polyphenylene ether and conductive carbon black. The obtained resin composition in the form of pellets was molded into test specimens, and the volume resistivity-2, deflection temperature under load and Izod impact strength of the resin composition were measured using the test specimens in the above-mentioned manner. The results are shown in Table 2, together with the formulation of the resin composition.

[0108]

[Comparative Example 4]

A resin composition was produced by a method described in patent document 3. Specifically, the resin composition was produced in substantially the same manner as in Comparative Example 3, except that masterbatch PA/KB-MB3 was used in an amount as shown in Table 2 below instead of the conductive carbon black, and the masterbatch was fed to the extruder from the 2nd downstream-side inlet. The properties of the resin composition are shown in Table 2, together with the formulation of the resin composition.

[0109]

[Examples 2 to 4]

In each of Examples 2 to 4, a conductive masterbatch in the form of pellets (master pellets) having a low surface luster was produced in substantially the same manner as in Com-

parative Example 2, except that the design of the screws of the twin-screw extruder (used for melt-kneading 90 parts by weight of polyamide 6 and 10 parts by weight of conductive carbon black at a cylinder temperature of 270 °C) was changed. Specifically, the production of the masterbatch was conducted while controlling the melt-kneading conditions (the rotation rate of the screws, the extrusion rate and the like) to obtain master pellets having a surface roughness Ra of 0.3 µm or more (the melt-kneading conditions employed in each of Examples 2 to 4 were mild as compared to those employed in Comparative Example 2). (The masterbatches produced in Examples 2, 3 and 4 are referred to as "PA/KB-MB4", "PA/KB-MB5" and "PA/KB-MB6", respectively). With respect to the obtained master pellets, the number of agglomerated particles contained therein and the surface roughness Ra of the flat portions thereof were measured. In Examples 2 to 4, resin compositions were individually produced in substantially the same manner as in Comparative Example 2, except that the above-obtained conductive masterbatches were used in accordance with the formulations shown in Table 2 below. The properties of the resin compositions are shown in Table 2, together with the formulations of the resin compositions.

[0110]

[Example 5]

A resin composition was produced in substantially the

same manner as in Comparative Example 3, except that masterbatch PA/KB-MB5 was fed to the extruder in an amount shown in Table 2 from the 2nd downstream-side inlet, instead of the conductive carbon black. The properties of the resin composition are shown in Table 2, together with the formulation of the resin composition.

[0111]

[Table 2]

Table 2

Feeding inlet of the extruder	Components of the resin composition	Units	Formulation of the resin composition						
			Comp. Ex. 2	Comp. Ex. 3	Comp. Ex. 4	Ex. 2	Ex. 3	Ex. 4	Ex. 5
Top	PPE-1		20	20	20	20	20	20	20
	PPE-3		14	14	14	14	14	14	14
	SEBS-1		8.5	8.5	8.5	8.5	8.5	8.5	8.5
	SEBS-2		2.5	2.5	2.5	2.5	2.5	2.5	2.5
Side 1	PA66		36	53	36	36	36	36	36
	PA/KB-MB3	Parts by weight	19						
	PA/KB-MB4			19					
	PA/KB-MB5				19				
	PA/KB-MB6					19			
	PA/KB-MB3						19		
	PA/KB-MB5							19	
	KB			1.9					
Results of measurements (with respect to each of masterbatches PA/KB-MB3, PA/KB-MB4, PA/KB-MB5 and PA/KB-MB6)									
Number of agglomerated particles	pieces	0	-	0	19	4	44	4	
Surface roughness Ra	μm	0.25	-	0.25	1.18	0.49	1.6	0.49	
Results of measurements (with respect to each of the produced resin compositions (PA66/PPE/KB's))									
Volume resistivity-2	Ω·cm	4 × 10 <sup>6</sup>	5 × 10 <sup>6</sup>	7 × 10 <sup>6</sup>	8 × 10 <sup>6</sup>	7 × 10 <sup>6</sup>	8 × 10 <sup>6</sup>	7 × 10 <sup>6</sup>	
HDR under a high load (1.82 MPa)	°C	125	124	125	129	128	130	129	
Izod impact strength	J/m	210	180	210	210	220	185	205	

[0112]

From Table 2, it is seen that by using the masterbatch in which the conductive carbon black is present in the form of agglomerated particles, the deflection temperature under load (or high load HDT) of a resin composition comprising a polyamide, a polyphenylene ether and conductive carbon black can be improved and a high conductivity can be achieved while maintaining the impact resistance. Further, none of the masterbatches produced in Examples 2 to 5 contained conductive carbon black in the form of an agglomerated particle having a major axis of 100  $\mu\text{m}$  or more. In addition, the melt flow rate (MFR: determined in accordance with ASTM D 1238, i.e., a flow rate of a molten resin per 10 minutes at 280 °C under a load of 5 kg) of each of the resin compositions produced in Examples 3 and 5 was measured. As a result, it was found that the MFR's of the resin compositions of Examples 3 and 5 were 24 g/10 min. and 19 g/10 min., respectively. From the above, it is apparent that a conductive resin composition which exhibits excellent conductivity, impact resistance, heat distortion resistance and melt-fluidity can be obtained by a method which comprises the following steps: (1) producing a conductive masterbatch comprising a polyamide and conductive carbon black, wherein a part of the conductive carbon black is present in the form of an agglomerated particle having a major axis of 20 to 100  $\mu\text{m}$ , and (2) adding the produced conductive masterbatch to a molten poly-

phenylene ether.

[0113]

[Comparative Example 5]

An automobile fender was produced by molding the conductive resin composition produced in Comparative Example 2, and the produced fender was subjected to an automobile coating process to thereby electrostatically coat the fender. As a result, it was found that the electrostatic coating property of the resultant fender was unsatisfactory, and that the fender suffered heat distortion.

[0114]

[Comparative Example 6]

An automobile fender was produced by molding the conductive resin composition produced in Comparative Example 3, and the produced fender was subjected to an automobile coating process to thereby electrostatically coat the fender. As a result, it was found that the electrostatically coated fender suffered a large amount of heat distortion, and that the impact resistance of the fender was unsatisfactory for practical use.

[0115]

[Example 6]

An automobile fender was produced by molding the conductive resin composition produced in Example 2, and the produced fender was subjected to an automobile coating process to thereby electrostatically coat the fender. The resultant elec-

trostatically coated fender produced using the resin composition of Example 2 exhibited only a very small amount of heat distortion, and that the electrostatic coating property was satisfactory. Further, the electrostatically coated fender had satisfactory impact resistance. Thus, the electrostatically coated fender could be advantageously put into practical use. The surface roughness of the fender was 0.65 µm and the fender had an excellent appearance even after the electrostatic coating (see Figs. 5 and 6).

[0116]

[Effect of the Invention]

The conductive resin composition of the present invention contains a masterbatch in which the conductive carbon black is present in the form of an agglomerated particle and, hence, has excellent impact resistance and excellent conductivity and is advantageous in that, when the resin composition is molded and the resultant molded article is then subjected to an electrostatic coating, the resin composition is unlikely to suffer heat distortion or a lowering of the impact resistance, wherein excellent coating can be formed on such a molded article due to the excellent conductivity of the resin composition.

Therefore, a molded article produced using the conductive resin composition of the present invention can be used in a wide variety of fields, such as electric and electronic parts, parts of office automation machines, automobile parts and other

mechanical parts. Especially, the conductive resin composition is very advantageous as a material for producing an automobile outer panel (especially an automobile fender).

[Brief Description of the Drawings]

[Fig. 1]

A schematic explanatory view of a cylindrical pellet of a conductive masterbatch, showing a perspective view of a cross-section of the pellet, which cross-section is observed to determine the number of agglomerated particles of conductive carbon black.

[Fig. 2]

A schematic explanatory view of a spherical pellet of a conductive masterbatch, showing a perspective view of a cross-section of the pellet, which cross-section is observed to determine the number of agglomerated particles of conductive carbon black.

[Fig. 3]

A schematic explanatory view of a pellet of a conductive masterbatch (PA/KB-MB1), showing a perspective view of a cross-section of the pellet, which cross-section is observed to determine the number of agglomerated particles of conductive carbon black.

[Fig. 4]

A schematic explanatory view of a conductive masterbatch (PA/KB-MB2), showing a perspective view of a cross-section of

the pellet, which cross-section is observed to determine the number of agglomerated particles of conductive carbon black.

[Fig. 5]

A schematic explanatory view of a part of an automobile body around a front wheel.

[Fig. 6]

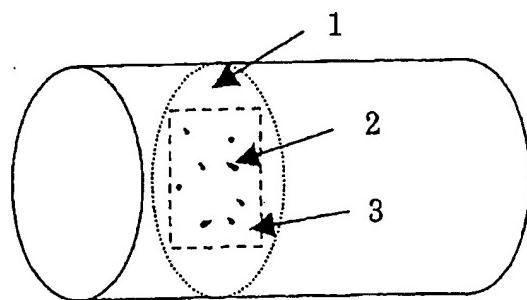
A schematic explanatory view of a part of an automobile fender; and an explanatory diagram showing a surface profile of a part of the fender.

[Description of Reference Numerals]

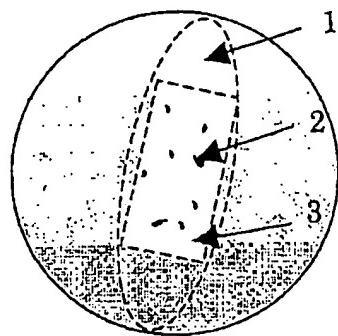
- 1: cross-section of a pellet
- 2: conductive carbon black
- 3: polyamide
- 4: diameter of a pellet
- 5: length of a pellet
- 6: central line average roughness Ra
- 7: fender of an automobile
- 8: outer surface of a fender ( $20 \mu\text{m} \times 20 \mu\text{m}$ )
- 9: expanded image of the outer surface of a fender (8)
- 10: explanatory diagram showing a surface profile
- 11: value of a surface roughness

[Name of Document] Drawing

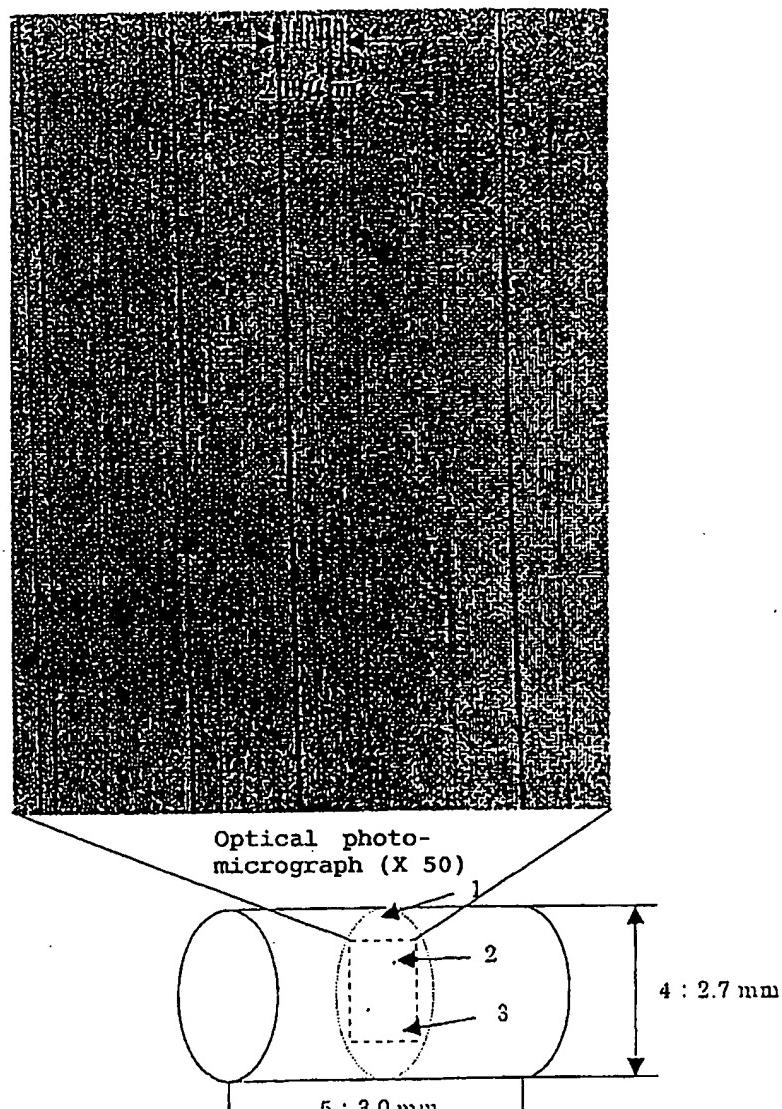
[Fig. 1]



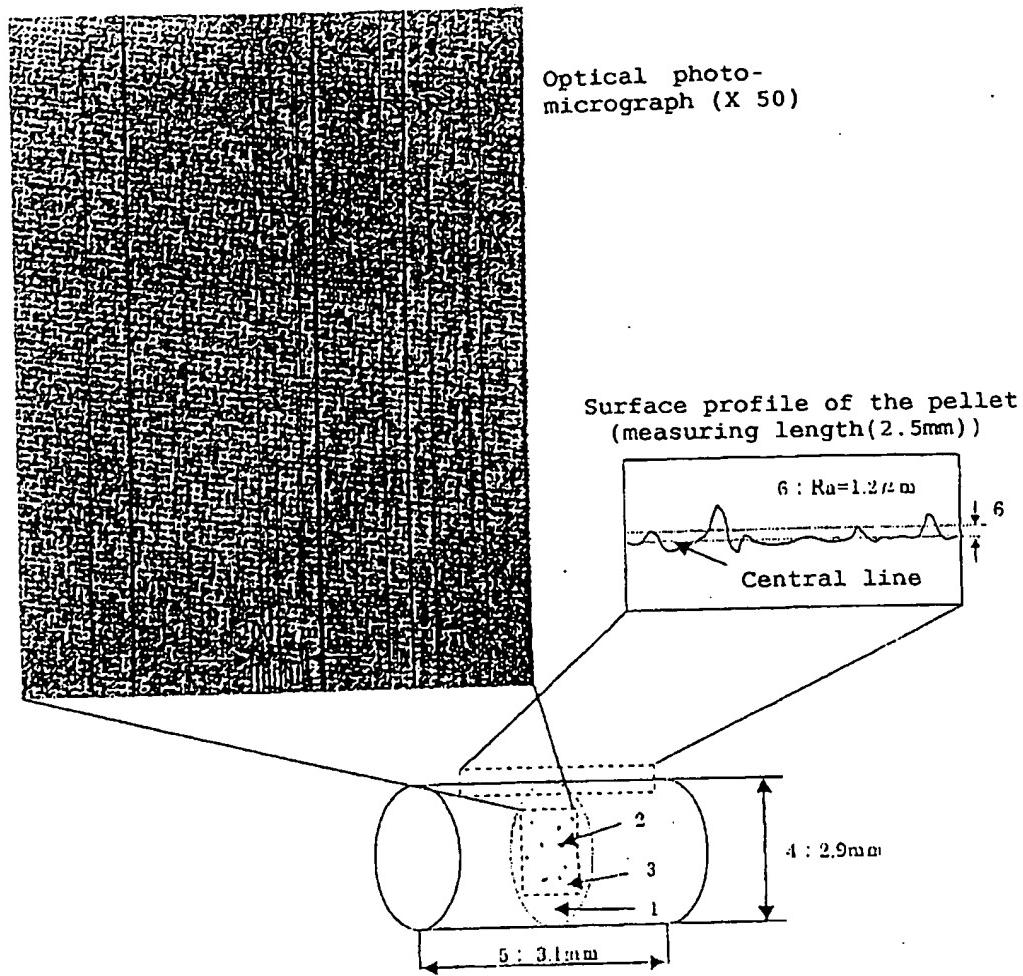
[Fig. 2]



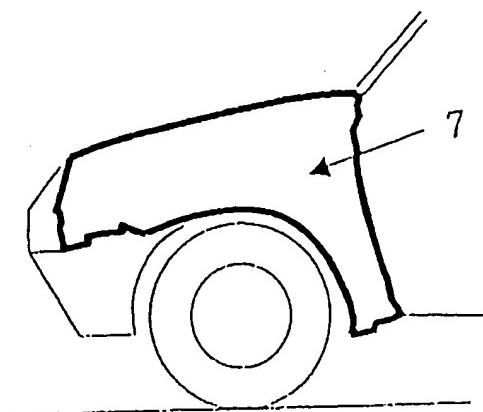
[Fig. 3]



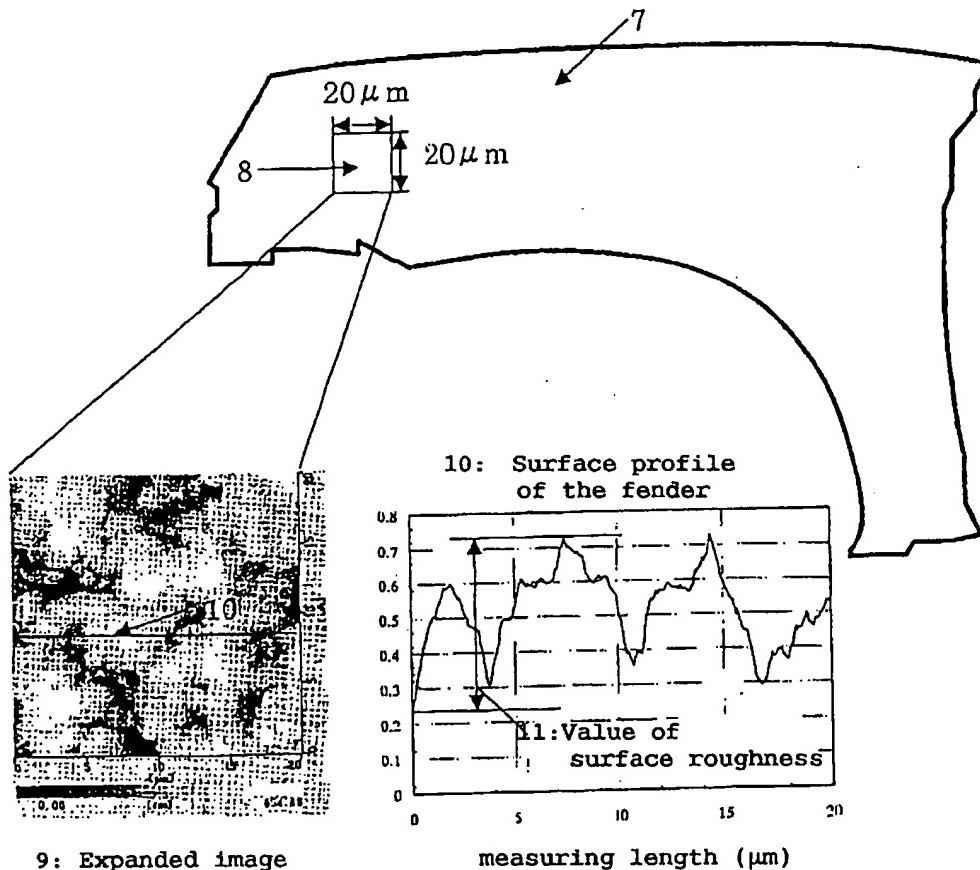
[Fig. 4]



[Fig. 5]



[Fig. 6]



[Name of Document] Abstract

[Abstract]

[Task] It is a task of the present invention to provide a masterbatch used for producing a resin composition having excellent impact resistance and excellent conductivity, which is advantageous in that, when the resin composition is molded into a large article (such as an automobile outer panel) and the resultant molded article is then subjected to an electrostatic coating, the resin composition is unlikely to suffer heat distortion and a lowering of the impact resistance, wherein excellent coating can be formed on such a molded article due to the excellent conductivity of the resin composition. It is another task of the present invention to provide a resin composition and molded article which are produced using the above-mentioned masterbatch; and a method for producing the above-mentioned resin composition or molded article.

[Means to Solve the Task] A method for producing a resin composition by adding a conductive masterbatch to a molten poly-phenylene ether, the conductive masterbatch comprising a polyamide and a conductive carbon black, wherein a part of the conductive carbon black is present in the form of 1 to 100 agglomerated particles each independently having a major axis of 20 to 100  $\mu\text{m}$ , as observed under an optical microscope with respect to a contiguous area of 3  $\text{mm}^2$ .

[Selected Figure] None

2003-153159

### Recognition • Additional Information

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2003-153159

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